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Artificial Healing of Cone Cracks in Glass

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Hertzian cone cracks formed by indenting brittle solids with a hard sphere have previously been shown to close imperfectly as the indenter load is released; accordingly, there is little tendency for air-formed cone cracks to heal naturally. In the present instance partial healing has been achieved artificially by subjecting residual cone cracks in glass to closure-enhancing treatments. Observations of the crack geometry both during and after loading suggest that healing occurs only at localised regions of intimate contact across the crack interface. Possible mechanisms for the healing process are discussed.

Es wurde schon früher gezeigt, daß Hertzsche Kegelsprünge, die durch Drücken eines spröden Festkörpers mit einer harten Kugel entstehen, sich nur unvollständig schließen, wenn die Stempellast gelöst wird; demgemäß besteht nur eine geringe natürliche Ausheiltendenz für an Luft gebildete Kegelsprünge. Im vorliegenden Falle wurde partielle, künstliche Ausheilung verbliebener Kegelsprünge in Glas durch eine den Verschluß fördernde Behandlung erreicht. Beobachtungen der Sprunggeometrie während und nach der Belastung weisen darauf hin, daß Ausheilung nur in lokalisierten Bereichen mit unmittelbarem Kontakt über die Sprungzwischenfläche stattfindet. Mögliche Mechanismen für den Ausheilprozeß werden diskutiert.

1. Introduction

In a recent article the closure of Hertzian cone cracks in brittle solids was reviewed [1]. As a result of the failure of complementary surface markings on opposing cone crack walls to key together exactly upon release of the crack-producing load, a residual crack interface remains in the test specimen. The mismatch of material across the interface is nevertheless small, typically measurable on an Ångstrom scale, suggesting that closure, and perhaps also healing, might be induced by artificial means. This leads to the possibility of modifying the structure of a residual fracture interface in a controlled manner. Such a possibility has implications in such diverse fields as material strength, semiconductor device fabrication, etc. [1].

In this paper we describe attempts to artificially heal residual cone cracks by a) applying compressive stresses across the crack interface, and b) subjecting the cracked specimens to a heat treatment.

2. Treatment of Residual Cone Cracks

Our experimental procedure follows that outlined previously [1]. A cone crack is produced in a glass slab by critically loading a steel ball on the glass surface. The formation and subsequent progress of the crack are followed during the indentation cycle by means of a microscope located beneath the specimen. With our arrangement the load on the indenter could be released and reapplied at will, and indenting spheres could be interchanged without disturbing alignment.

From theoretical considerations it can be argued that the degree of healing that occurs at a residual fracture interface should be reflected by the progress of the cone crack during a subsequent reloading stage with the original indenter [1]. In particular, if a critical load is again required to re-initiate the original cone crack then at least partial healing must have occurred. No such behaviour was observed in the previous examination of several hundred cone cracks reloaded directly after the first loading cycle: in all cases the fracture interface reappeared on immediate reapplication of the load.

In the present instance we have subjected freshly formed cone cracks to closure-enhancing treatments prior to the reloading cycle.

2.1 Stress-enhanced closure

In applying a compressive stress to the residual crack interface use is made of the large component of hydrostatic compression within the Hertzian stress field itself [2]. Within a drop-shaped zone beneath the contact area all three principal stresses are compressive, the hydrostatic pressure rising to a maximum $\approx 1.3\, p_0$ (p_0 being the mean indentation pressure) at the centre of contact. Outside this zone where the cone crack ultimately forms the greatest principal stress becomes tensile, reaching a maximum of $\approx 0.16\, p_0$ at the contact circle. The indentation pressure is measurable in terms of the size of the contact circle [3]:

$$p_0 = \left(\frac{3\,E}{4\,\pi\,k}\right)\frac{a}{r}, \tag{1}$$

where a is the radius of the contact circle, E is Young's modulus, k is a dimensionless constant of order unity, and r is the radius of the indenter. Equation (1), although strictly based on an elastic contact, conveniently holds even after the cone crack is formed [1].

The particular sequence of events adopted is shown in Fig. 1:

a) A 1/2 in. thick glass slab is loaded with a small steel ball (radius $r = 1/8$ in.) so as to produce a cone crack.

b) The indenter load is released, allowing the crack walls to move together. (The rim of the residual crack is usually faintly visible at this stage.)

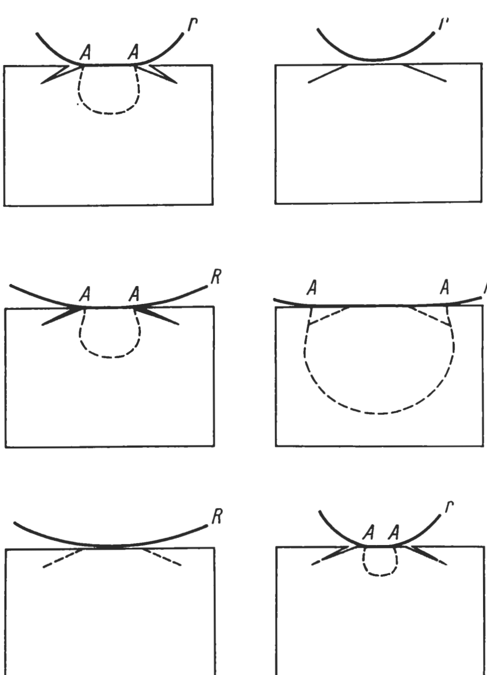

Fig. 1. Sequence for closing cone cracks. Drop-shaped compressive zone indicated beneath contact circle AA. r and R denote radii of small and large indenters respectively

c) A larger ball (radius $R = 3/4$ in.) is loaded along the symmetry axis of the residual cone crack. The crack suffers an increasing tension up to the point at which the surface rim becomes engulfed within the compressive zone. At this point the contact geometry is similar to that in stage a), with the stress level correspondingly lower by a factor of $r/R = 1/6$ (holding a constant in equation (1)). Thus there is only a slight tendency for the larger indenter to reopen the residual cone crack.

d) The larger ball is loaded further, until the compressive zone fully encompasses the entire cone crack. This occurs when the diameter of the contact is approximately twice the diameter of the surface trace of the cone crack. Thus the stress level at stage d) is (according to equation (1)) approximately twice that at stage c). Bearing in mind the order of magnitude difference between values of compressive stresses within the drop-shaped zone and tensile stresses outside it, it may be appreciated that the residual crack is subjected to a relatively substantial compression.

e) After maintaining the pressure as in d) for a certain time, the larger ball is removed.

f) The original, smaller ball is reapplied to the crack, and the regrowth characteristics of the cone crack noted.

2.2 Temperature-enhanced closure

Cone cracks are produced as in Fig. 1a and b above. The specimens are then annealed in a nitrogen atmosphere for a certain time. The residual cracks are then subjected to a reloading cycle, as in Fig. 1f.

3. Behaviour of Treated Cone Cracks

3.1 Observations of crack opening during loading cycle

As pointed out previously [1] the progress of the *tips* of the cone cracks is difficult to follow optically, owing to the fine scale of the crack-wall separation. The closure and healing characteristics could, however, be inferred from the Fizeau fringe system generated by critically reflecting sodium light from the crack interface into the microscope below the specimen [1]. With the 1/8-in. radius indenter at critical load (Fig. 1a) three fringes were usually observed, indicating a crack-mouth opening of nearly one micron. Throughout the indenting procedure the fringe positions were recorded in terms of the radial distance $\mathscr{R}$ measured in the image plane (Fig. 2).

From such optical observations some distinctive evidence for crack "healing" was obtained for both compressed and heated specimens. Favourable cases showed a sudden reappearance of one or more fringes during reloading. The results varied considerably according to such parameters as indenter load rate, critical indenter load for the original cone crack formation, and duration and degree of the compression or anneal treatment. The first of these enters as a parameter because the test environment influences the Hertzian strength of glass in a rate-dependent manner [4]: a meaningful comparison between crack behaviour before and after treatment thus required standardisation of the load rate. The second arises as a result of the wide range of Hertzian strength values characteristic of commercial glass slabs, cracks initiated at lower critical loads showing a greater tendency to grow without complication: a light abrasion of the glass surfaces was sufficient to give consistently low critical loads [5]. In

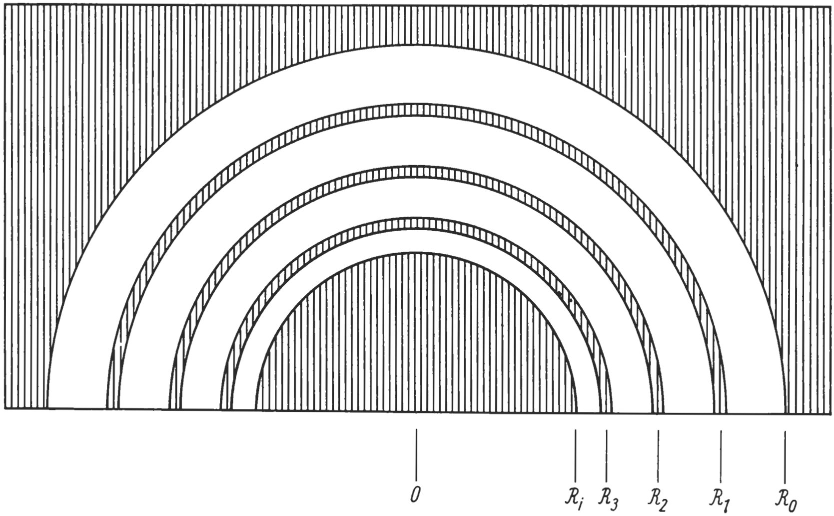

Fig. 2. Cone crack Fizeau fringe system in (half) image plane. $\mathcal{R}_i$ and $\mathcal{R}_0$ designate inner and outer radii of crack mage (actual crack tip lies somewhere outside $\mathcal{R}_0$). $\mathcal{R}_m$ ($m = 1, 2, 3 \ldots$) designates m-th order dark fringe. (The radius a_c of contact circle at critical loading, which lies within $\mathcal{R}_i$, is measured in normally reflected light)

controlling these first two parameters, the effects of the closure-enhancing treatment could be followed systematically.

Fig. 3 indicates the movement of the Fizeau fringe system for a well-healed cone crack. The observed sequence of events is as follows: (i) The indenter loads elastically along stage 1 until the contact circle reaches a critical value. (ii) The cone crack quickly grows along stage 2. (iii) On unloading the indenter the fringe minima contract, while the crack tip region remains stationary along stage 3. (iv) The residual crack is "closed" artificially (in this case by compression) along stage 4. (v) The crack is reloaded with the original indenter along stage 5. (vi) At a second critical contact radius (lower than the first) the crack suddenly reappears along stage 6. (vii) On subsequent further loading and unloading along stages 7 and 8 the fringe system retraces the first unloading stage 3.

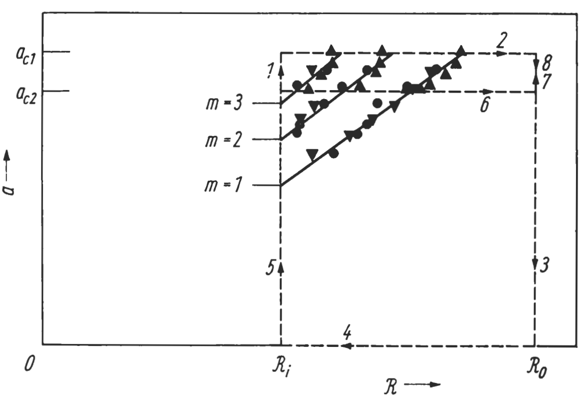

Fig. 3. Plot showing expansion and regression of Fizeau fringe system during loading cycles. Circles denote regression of fringe minima during unloading stage 3. Upright triangles denote expansion of fringes during reloading stage 7, and inverted triangles the regression during unloading stage 8, Crack compressed during stage 4 for 105 min. at $a_{comp}/a_{c1} = 2.9$

3.2 Effect of variations in crack treatment

By simply recording the critical contact radii as in Fig. 3 (stages 2 and 6), a convenient "healing parameter",

$$H = \frac{a_{c2}}{a_{c1}} \tag{2}$$

could be readily evaluated for each treated cone crack. Thus $H = 0$ indicates zero healing, $H = 1$ perfect healing.[1])

In Fig. 4 the healing parameter is plotted as a function of cone crack compression duration, for various compression factors a_{comp}/a_{c1}, where a_{comp} is the radius of the contact circle for the compressing indenter (Fig. 1d). H is seen to increase slightly with both time and degree of compression. In Table 1 the healing parameter is listed for different annealing times and temperatures. In this case the dependence of H on the variables of the closure-enhancing treatment is less systematic.

Table 1

Healing parameter for several anneal times and temperatures. Three results per specimen. (Dash indicates no healing)

Anneal temp. (°C)	Anneal time (approx.)	Healing parameter H		
200	1 hour	—	0.87	0.79
400	1 hour	0.82	0.79	—
400	1 day	0.64	0.83	—
400	1 week	0.94	0.96	0.88
600	1 week	0.94	0.88	0.97

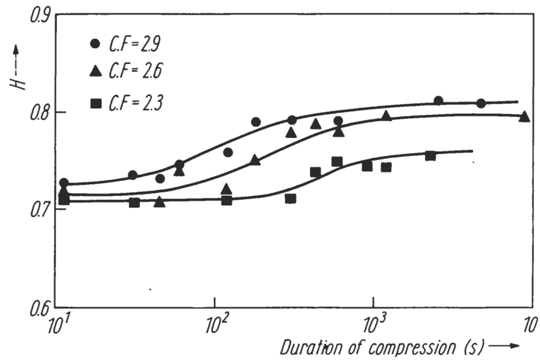

Fig. 4. Healing parameter H as function of compression time, for several compression factors C.F. $= a_{comp}/a_{c1}$

[1]) For intermediate values H should be considered as no more than a figure of merit. The healing parameter could, for instance, have been equally well defined in terms of the critical indenter loads P_{c1}, P_{c2} rather than the contact radii. According to Hertzian contact theory $P \sim a^3$ [1], so that the alternative definition would give smaller values of H within the above limits, and would also be more sensitive to small changes in the healing. Moreover, P_c represents a measure of the fracture surface energy of the solid [4], and the alternative definition would therefore relate more directly to the strength of the fracture interface. Nevertheless, we will be concerned only with trends in healing behaviour here, for which purpose the definition (2) above will suffice.

3.3 Observations of residual cracks

Despite the *healing* characteristics shown by the treated cone cracks, a closer examination of the residual interface indicated that *closure*, while always enhanced, was never complete.

First, two-beam interference measurements of the surface distortion around residual cracks [6] on unabraded specimens indicated typical reductions in residual mouth opening from about a quarter to a tenth of a fringe spacing (sodium light) as a result of the closure-enhancing treatment. This suggests that the healing process occurs only at localised "high spots" of intimate contact across the crack interface.

Second, cracked specimens were sectioned normal to the indented surface down to the cone-crack diameters and etched in 5% HF for thirty seconds. Simultaneous examination of adjacent untreated and treated cracks on the same section showed no detectable differences in etch pattern: a completely closed crack might have been expected to etch less vigourously. In the earlier study of untreated cracks [1] it was found that the crack etches over the entire optically detectable length (as observed in Section 3.1). Thus even the best-healed residual cracks represent interfaces of high energy in the solid.

4. Discussion

That glass surfaces in intimate contact are capable of showing mutual adhesion was first demonstrated by Lord Rayleigh [7]; two clean, dust-free optical flats pressed together (in air) required substantial forces to separate them. The situation at the residual fracture interface is not dissimilar. A compressive force applied to the crack will inevitably restore contact over at least a portion of the interface, and this contact is likely to increase with the magnitude of the force. It should be pointed out that even when the externally applied compression is zero there exist internal driving forces tending to bring opposing crack walls into contact: first, the residual stresses associated with an unclosed crack can vanish only upon perfect closure, thereby giving rise to a restoring force approximately proportional to the crack-wall separation; second, cohesive forces operating across the fracture interface close to the crack tip act to pull the crack walls together. These restoring forces appear to be too small to induce significant natural closure and healing of freshly formed cracks in glass at room temperature [1].

Without detailed observations of the structure of the artificially closed cone crack interfaces it is not easy to establish the mechanism of the induced healing. In view of the lateral mismatch that exists across the residual crack interface [1] it is to be expected that recontact should occur predominantly at localised structural prominences on the cleavage faces, in accordance with the observations of Section 3.3. At such localised regions the contact stresses would be highly concentrated. In materials which deform readily in a plastic manner at room temperature, e.g. metals [8] and alkali halide crystals [8, 9], such stresses are sufficient to form "plastic junctions"; in this way fresh surfaces are brought into "true contact", and adhesion arises as a result of atomic rebonding [8]. Strong adhesion also occurs between cleavage surfaces of dielectric minerals, such as mica [10 to 12, 8] and certain silicates [13 to 15]. When tested in ultrahigh vacuum there is evidence for considerable electrostatic charging of silicate cleavage surfaces; long-range attractions between the surfaces, substantial for even millimeter-scale separations, have been observed [14]. In air, however, the surfaces become contaminated, and the adhesion is reduced by at least an order of magnitude. In these cases the charge and coordination requirements of exposed bonds on a freshly cleaved surface are certain to be satisfied by the adsorbed species, and the adhesive bonds acting across the interface are likely to be of the weak, secondary type, e.g. van der Waals and hydrogen bonds. Nevertheless, even air-cleaved specimens may, under certain conditions, show evidence for localised restoration of the original primary bonds [13]. Finally, at temperatures at which diffusion processes become significant, crack healing may take place as the result of matter transport to the tip region; the kinetics of such processes have been studied in ionic crystals at elevated temperatures [16].

Recently Wiederhorn and Townsend put forward evidence for spontaneous healing of cracks in double-cantilever specimens of glass [17]. They found that healing was enhanced by forming the fractures rapidly. This time-dependent effect is in accord with the well-known static fatigue behaviour of glass: the weakening effect of environmental interaction with the crack faces is reduced as the rate of loading the specimen is increased [4]. On the basis of this observation and recent studies of the bond structure at glass surfaces (other workers), Wiederhorn and Townsend concluded that the adhesion occurs via primary bond restoration for quickly-formed (relatively "clean") fractures, and via secondary bond interaction for slowly-formed (contaminated) fractures. The observed behaviour of the double-cantilever cracks does not, however, appear to be completely analogous to that of cone cracks: neither impact- nor pressure-formed cone cracks show any tendency to heal naturally [1].

Any description of the artificially healed cone crack interfaces in the present instance must account for the time-dependent effects observed (e.g. Fig. 4). The appearance of a time factor in the mechanical behaviour of glass is, in fact, not uncommon. Apart from the rate-controlled *surface* interaction processes which give rise to the fatigue behaviour mentioned above, the rearrangement of the *bulk* structure of glass around residual strain centres (e.g. Vickers hardness impressions [18]) leads to pronounced recovery effects. Thus, while from the previous discussion we might have predicted a healed interface in which secondary bonding is predominant for the air-formed cracks, the possibility of molecular rearrangement leading to primary re-bonding at points of intimate contact (in analogy to the formation of plastic junctions in metals) can not be completely discounted.

5. Conclusion

It has been shown that healing can be artificially induced in air-formed cone cracks in glass. Because the stresses required to re-open the cone crack are somewhat weaker than those originally required to form it, it is concluded that the healed residual crack represents a weakly-bonded interface in the solid. More systematic experiments in ultra-high vacuum, and in controlled environments, would no doubt help to clarify the nature of the healing mechanisms.

Finally, since the geometry of the cone crack is likely to resemble that of surface flaws introduced into brittle solids by handling, etc., studies of the type outlined above are directly relevant to a description of material strength. To point out one example, Mould [19] found that both vacuum baking and ageing (in several environments) of freshly-abraded glass slides led to an increase in the fracture strength of the glass. Not being able to observe directly the behaviour of the individual abrasion-induced flaws responsible for the fracture, Mould could not make a definitive statement concerning the strengthening mechanism. Although he did not favour a healing concept, the present evidence suggests that this possibility warrants further consideration.

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References

[1] J. S. Williams, B. R. Lawn, and M. V. Swain, phys. stat. sol. (a) **2**, 7 (1970).
[2] B. R. Lawn, J. appl. Phys. **39**, 4828 (1968).
[3] M. V. Swain and B. R. Lawn, phys. stat. sol. **35**, 909 (1969).
[4] F. B. Langitan and B. R. Lawn, J. appl. Phys. **41**, 3357 (1970).
[5] F. B. Langitan and B. R. Lawn, J. appl. Phys. **40**, 4009 (1969).
[6] B. R. Lawn and H. Komatsu, Phil. Mag. **14**, 689 (1966).
[7] Lord Rayleigh, Nature (London) **139**, 781 (1937).
[8] F. P. Bowden and D. Tabor, The Friction and Lubrication of Solids, Part II, Oxford University Press, 1964.
[9] A. J. Forty and C. T. Forwood, Trans. Brit. Ceram. Soc. **62**, 715 (1963).
[10] J. W. Obreimoff, Proc. Roy. Soc. **A127**, 290 (1930).
[11] E. Orowan, Z. Phys. **82**, 239 (1933); **82**, 259 (1933).
[12] P. J. Bryant, L. H. Taylor, and P. L. Gutshall, Trans. 10th National Vacuum Symp., MacMillan Co., New York 1963 (p. 21).
[13] J. A. Ryan, J. geophys. Res. **71**, 4413 (1966).
[14] J. A. Ryan, J. J. Grossman, and W. M. Hansen, J. geophys. Res. **73**, 6061 (1968).
[15] J. A. Ryan, and J. J. Grossman, Science J. **4**, 41 (1968).
[16] Ya. E. Geguzin, N. N. Ovcharenko, and I. V. Vorobeva, Soviet Phys. — Cryst. **10**, 299 (1965).
[17] S. M. Wiederhorn and P. R. Townsend, J. Amer. Ceram. Soc. (in the press).
[18] W. Diehl and R. Schulze, Z. angew. Phys. **9**, 251 (1957).
[19] R. E. Mould, J. Amer. Ceram. Soc. **43**, 160 (1960).

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